Supporting Information for

A Hydrocarbon Cathode for Dual-Ion Batteries

Ismael A. Rodríguez-Pérez, Zelang Jian, Pieter K. Waldenmaier, Joseph W. Palmisano, Raghu Subash Chandrabose, Xingfeng Wang, Michael M. Learner, Rich G. Carter & Xiulei Ji**

Figure S1. Scanning electron microscopy imaging. **a)** SEM image of a pristine coronene electrode showing a clear image of the coronene crystals. **b)** SEM image of the coronene electrode once fully charged. It is evident that upon charging, the morphology of the crystals has changed, showing a cracked, porous structure. **c)** SEM image of coronene electrodes after 400 cycles showing a completely amorphous structure.

*E-mail: rich.carter@oregonstate.edu
E-mail: david.ji@oregonstate.edu

Figure S2. Cut-off voltage optimization. Galvanostatic charge/discharge profiles of the first cycle with cut-off voltages of 4.3, 4.4, and 4.5 V. All tests showing extremely small coulombic efficiencies.

Figure S3. PF_6^- insertion site. Schematic of the possible site of a PF_6^- anion upon charging of the coronene electrode. The PF_6^- anion is in between the (111) (blue) and (200) (green) coronene planes, causing an expansion in the (a) direction.

Figure S4. *Ex situ* NMR measurements. The peak at ~ 8.75 ppm is the main and most important peak representative of pure coronene that is the emphasis of these spectra (NMR not shown due to a single peak present; ^1H NMR (700 MHz, C_6D_6) δ 8.75 (s, 12H)), the rest are due to solvent and partial impurities, including the binder used to make the electrode. **a)** NMR spectrum of the electrode before cycling (pristine); ^1H NMR (700 MHz, C_6D_6) δ 8.75 (s, 12H), 5.52 (s, 2H), 2.90 (s, 1H), 2.12 (t, $J = 0.7$ Hz, 2H), 1.40 – 1.21 (m, 20H), 0.89 (s, 2H) ppm. **b)** NMR spectrum of the electrode after a full charge. These spectra remained the same for all other *ex situ* measurements, hence omitted; ^1H NMR (700 MHz, C_6D_6) δ 8.75(s, 12H), 5.51 (s, 1H), 3.19 (s, 3H), 2.92 (s, 17H), 2.12 (t, $J = 0.7$ Hz, 2H), 1.73 (t, $J = 0.7$, 1H), 1.40 – 1.21 (m, 6H), 1.01 (s, 2H) 0.89 (s, 1H) ppm.

Figure S5. Long cycling profiles. Charge/discharge potential profiles for the 50th, 100th, 200th, 300th, and 400th cycles showing instrumental evidence of structural changes by 50 cycles; however, these structural changes lead to stable cycling.

Figure S6. X-ray patterns of structural evolution. X-ray diffraction patterns of the 2nd, 3rd, 50th, and 200th cycles. It is noticeable that by the 50th cycle, we start to see a significant structural change due to peak fading. Furthermore, a broad peak starts to be noticeable, indicating the crystal structure is turning slightly amorphous. Upon further cycling (200 cycles), the coronene has undergone severe structural changes and has lost all crystallinity. This evidence of structural changes coincides with the charge/discharge curves in Figure S6, where the previously seen plateaus (first 10 cycles in Figure 4a) are completely turned into a sloping region by the 50th cycle.